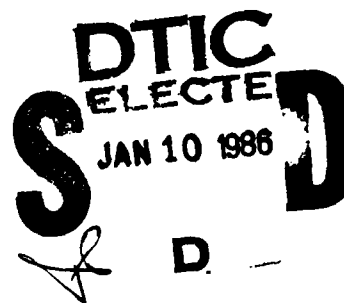**AD-A163 140**

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Final Technical Report
September 1985

PHOTOCHROMIC MATERIALS STUDY

San Diego State University

Dr. Jeffrey A. Davis and Mary Thomas

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19 ABSTRACT (Continue on reverse if necessary and identify by block number)
This report describes a study conducted to determine the feasibility of using the photochromic material mercury dithizonate as a programmable spatial filter for optical processing. This material has exhibited faster sriting speed and finer resolution than conventional spatial filtering techniques. Photochromics are erasable and therefore reusable. Variations in material performance were discovered and are attributed to the presence of two photochromic species in the material. A follow-on effort will investigate the effects of varying the composition of the photochromic material upon its optical properties.

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TABLE OF CONTENTS

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I. INTRODUCTION

We have been studying the photochromic material mercury dithizonate for use in optical signal processing applications under contract No. F320602-84-K-0110.

To briefly summarize, we have found a photochromic material (mercury dithizonate) whose optical transmission at red wavelengths decreases when the sample is illuminated with light in the blue-green spectral region. This material can therefore serve as an optical recording medium. Moreover, since the photochromic molecules relax to their unexcited state when the exciting light is turned off, this recording medium can be reused. Therefore, this material can serve as a reusable optical recording medium for use in a series of optical data processing applications.

Although previous measurements on mercury dithizonate have been reported, the high writing energies required to make the samples opaque reduced interest in this material. However, we felt that the development of argon lasers capable of high output powers in the blue-green spectral region should significantly reduce the time required to write on this material and therefore stimulate additional interest in these materials. Specifically, we are interested in decreasing the writing time in order to make it operate as a very fast, inexpensive, reusable recording medium.

Therefore, we have investigated both the writing and relaxation characteristics of these photochrpmic films as functions of writing beam intensity, writing wavelength, sample temperature, and reading beam intensity. Most of our results have been detailed in the individual progress reports which have been submitted. This report will basically review those conclusions with graphs as needed.

1

II. EXPERIMENTAL DETAILS

As mentioned earlier, both the writing and erasure
characteristics of the photochromic material were studied.

In the writing experiments, the sample was illuminated with light
from a Coherent model 90-3 high power argon laser at either the
514.5 nm or the 488.0 nm wavelength. This beam is referred to as
the writing beam. In addition, the sample was also illuminated
with light from a helium neon laser at a wavelength of 632.8 nm
which is the reading beam.

The writing experiments involved measurement of the transmission
of the sample at the helium neon laser wavelength as a function
of time during the writing phase. In these experiments, the
transmission of the sample would decrease as a function of time.

In addition, the relaxation or decay mechanism was studied by
following the increase in the transmission of the sample after
the writing beam was terminated.

A rather sophisticated data analysis system was used in both
kinds of experiments. The transmitted light intensity at the
wavelength of 632.8 nm was recorded as a function of time using a
Nicolet digital oscilloscope. The data was transferred to an
Apple computer for analysis and plotting.

A central thesis of our research is that a certain exposure
energy is required to make the photochromic material opaque and
that the exposure time needed to write the photochromic to a
certain optical density can be decreased by increasing the
intensity of the writing beam.

In our initial experiments, the illumination time was controlled
with a simple photographic shutter. However to avoid sample
damage when using higher illumination intensities, we had to
introduce an acousto-optic light modulator to decrease the
writing time.

The AOLM works by having an acoustic wave act as a diffraction
grating for the optical beam. Therefore, high diffraction
efficiency depends on having the acoustic wave completely overlap
the optical beam. Since the acoustic wave travels with the
velocity of sound, the risetime of the optical beam is limited by
the transit time of the acoustic beam through the optical beam.
This can be minimized by making the diameter of the optical beam
smaller. However, the higher power density resulting from the
focused beam can destroy the AOLM crystal. Using our system,
risetimes of about 200 nsec were obtained giving minimum pulse
widths of about 500 nsec.

Initial experiments were carried out by simply expanding both

laser beams to achieve uniform but low illumination intensities.
Higher writing intensities were obtained using the unfocused
argon laser beam as it came directly from the laser. This
presented a problem since the variation in argon laser intensitiy
over the beam profile results in a different writing speed for
different portions of the spot. To insure that the area probed
by the helium neon laser beam was uniformly illuminated, the
helium neon laser beam was focused so its total beam probed an
area of constant argon laser intensity. An EG&G Reticon diode
array was used to measure the profiles of both beams and Figure 1
shows typical experimental results. The outer curve shows the
intensity as a function of radius for the argon laser beam
profile while the center curve shows the helium neon laser
profile. The noise on the curves reflects the lack of uniformity
in the relative response of the elements in the photodiode array.

Higher illumination intensities were obtained by focusing both
beams. However, the alignment difficulties in making both beams
overlap at the sample surface as well as the uncertainties in
measuring the exact writing intensity can introduce experimental
errors.

The sample temperature is controlled with a MMR model K770T Joule
Thompson refrigerator system capable of temperatures from -197 to
+100 degrees C. The unit allows optical transmission experiments
and provides temperature stability of 0.1 degrees C.

Several experimental uncertainties limited our highest optical
power measurements. The focused spot size was smeared due to
heating of the acousto-optic light modulator from the high power
laser beam. We had difficulties in precisely aligning the two
focused laser beams onto the surface of the photochromic sample.
Finally, the uneven surfaces and different thicknesses of the
photochromic films affected the location of the laser focal spots
relative to the sample surfaces.

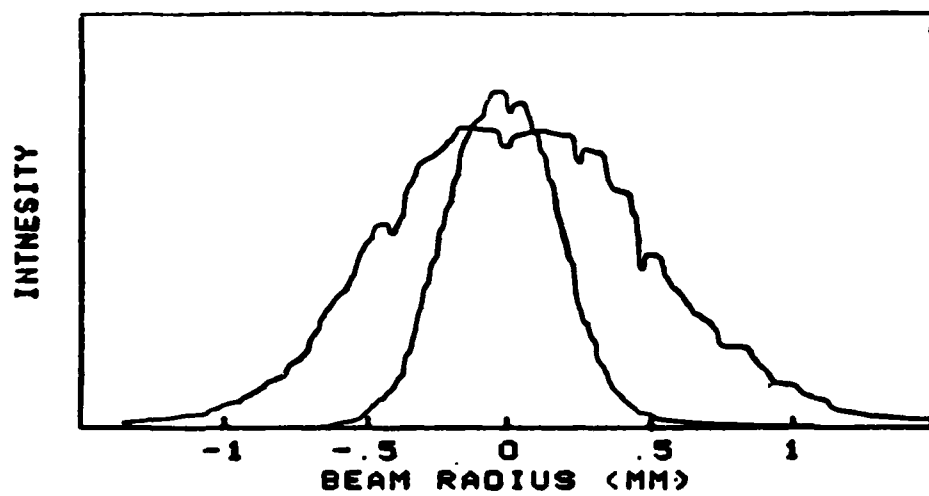Next, the decay time measurements will be discussed.

Figure 1 - Beam profiles of the argon and helium neon laser beams at the plane of the photochromic sample showing their relative positions and sizes for a low power argon beam.

4

III. DECAY RATE EXPERIMENTS

Our first series of experiments examined the decay or relaxation
rate as a function of temperature. We found a dramatic decrease
in the relaxation time as the sample temperature is increased and
the results are shown in Figures 2 - 5.

Figure 2 shows the relative transmission of sample #5 at a sample
temperature of 20 degrees C as a function of time after the argon
writing beam was turned off. This figure shows the long decay
time needed for the sample to return to the original transmitting
state.

Figure 3 shows the same experiment done at a temperature of 70
degrees C. The time scales are kept the same in the two figures
for ease of comparison. Here the sample recovers much more
quickly. The signal noise is due to fluctions in the helium neon
laser output power and was normalized out in later experiments.

Figure 4 shows the natural log of the decay time (in seconds) as
a function of temperature assuming a single exponential time
constant. This graph is qualitatively the same as shown in our
original proposal. However, we have extended the temperature
range. Extending the curve in Figure 4 shows that the erasure
time would approach two minutes at 100 degrees C and can be
shortened by increasing the sample temperature further. However,
at some point the sample will melt.

If we assume that some activation energy is required to convert
the excited molecule in its opaque state into the transparent
state, then the decay time should follow an Arrhenius Equation:

$$\ln (\Upsilon) \approx -E/kT$$

where E is the activation energy, k is Boltzman's constant, and
T is the sample temperature in degrees Kelvin.

Accordingly, we plotted the data as ln (Υ) versus 1/T as shown
in Figure 5. The slope of the straight line appears to justify
our assumption and gives a value of 0.45 eV for the activation
energy corresponding to a wavelength of about 2.8 microns.
Previously published (1) data shows an absorption peak in the
infrared spectrum of excited mercury dithizonate when in liquid
form at about 3 microns.

Measurements were carried out for our other samples and several
complicating factors arose. We have three different samples of
the photochromic material in plastic substrates and the decay
rate was studied as a function of sample temperature for each of
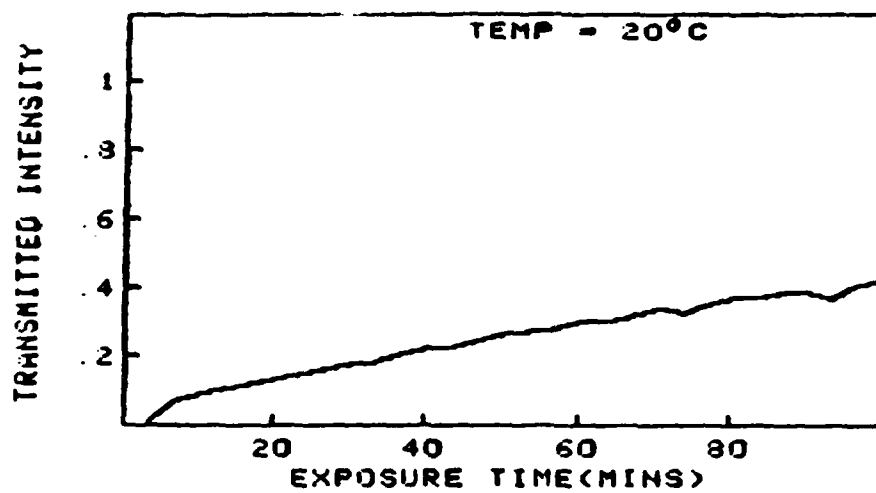these samples.

5

Figure 2 - Transmission versus time for sample #5 at a temperature of 20 deg C
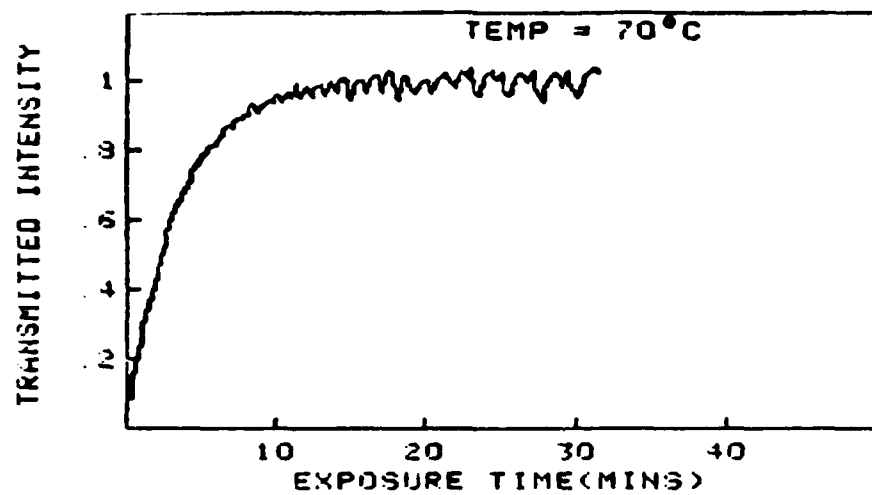
6

Figure 3 - Transmission versus time for sample #5 at a
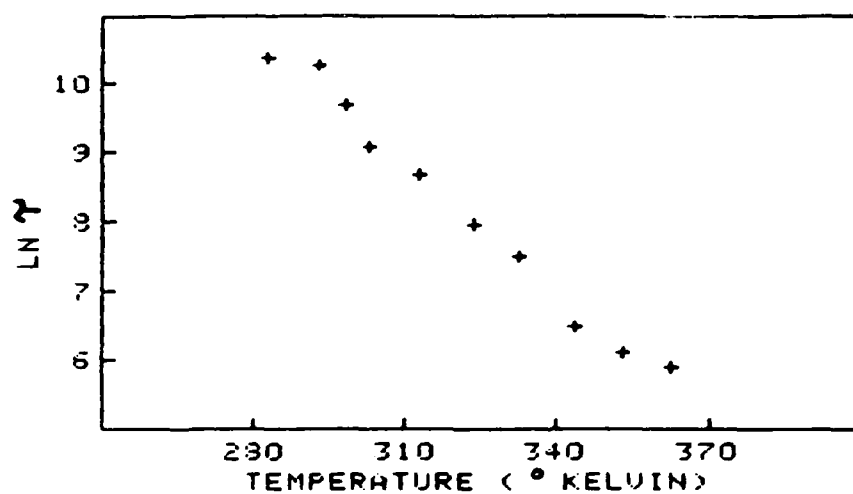temperature of 70 deg C

7

Figure 4 - Natural log of decay time (in seconds) versus
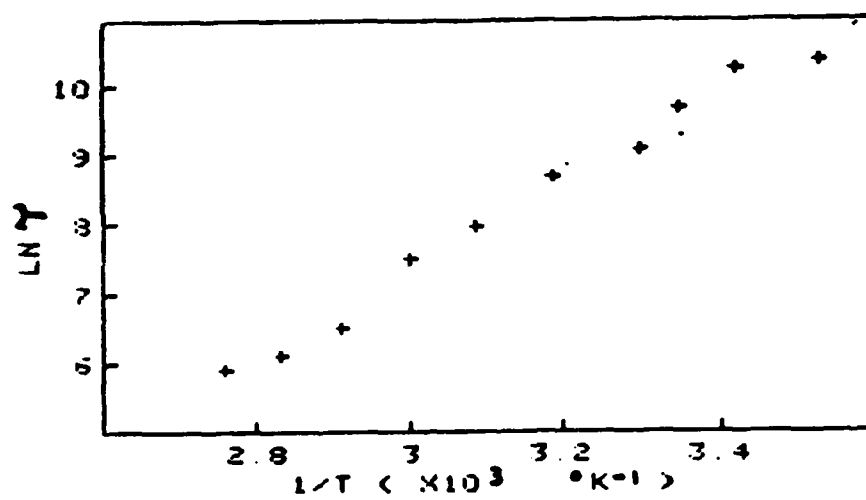temperature for sample #5

8

Figure 5 - Natural log of decay time (in seconds) versus inverse
temperature for sample #5

9

One sample (#5) was obtained three years ago from American
Optical Inc. Unfortunately, the person who made them has passed
away and we have no record concerning its composition.
Therefore, this sample is mainly used for initial measurements
and for troubleshooting purposes. The other two (#1 and #3) were
made by Dr. Nori Chu at that company. These samples have
different concentrations of the photochromic material.

All three samples show qualitatively the same behavior in that
their decay rate as measured at the helium neon laser wavelength
becomes faster as the sample temperature is increased.

However, we found evidence for two different decay times
governing the relaxation of the different photochromic samples.
At high sample temperatures of 90 degrees C, we find that
although the sample recovers the majority of its optical
transmission within a short time, a residual opacity remains
which decays with an extremely long time constant.

The primary decay behaves as reported earlier and is strongly
temperature dependent and, at high temperatures at least,
accounts for about 80% of the return to the transparent state.
We will next discuss the experimental results.

Figure 6 gives the decay as a function of time for sample #5 at
an argon laser intensity of 40 milliwatts/sq cm. If a single
decay constant were involved, this should yield a straight line
plot. It is this absence of a straight line which leads us to
postulate a two component decay rate.

Moreover, we discovered another complication. The decay rate was
accelerated when the helium neon reading intensity was increased.

Similar results were found for sample #1 as shown in Figure 7.
The decay is much slower and is also accelerated with increased
helium neon reading intensity.

Measurements of sample #3 were also carried out and are shown in
Figure 8. This sample shows a longer decay time than the others
and, in addition, shows the OPPOSITE behavior with increased
helium neon laser intensity.

Therefore, we see that all three samples behave differently, each
having a different overall decay rate and a different
susceptibility to the helium neon reading laser intensity.

Measurements of this effect are complicated by two factors.
First, drift in the laser monitor output power (in the 5% range
over periods of hours) increases the difficulty of making these
measurements. We therefore began recording the laser power at
the same time as the transmitted power through the sample, in
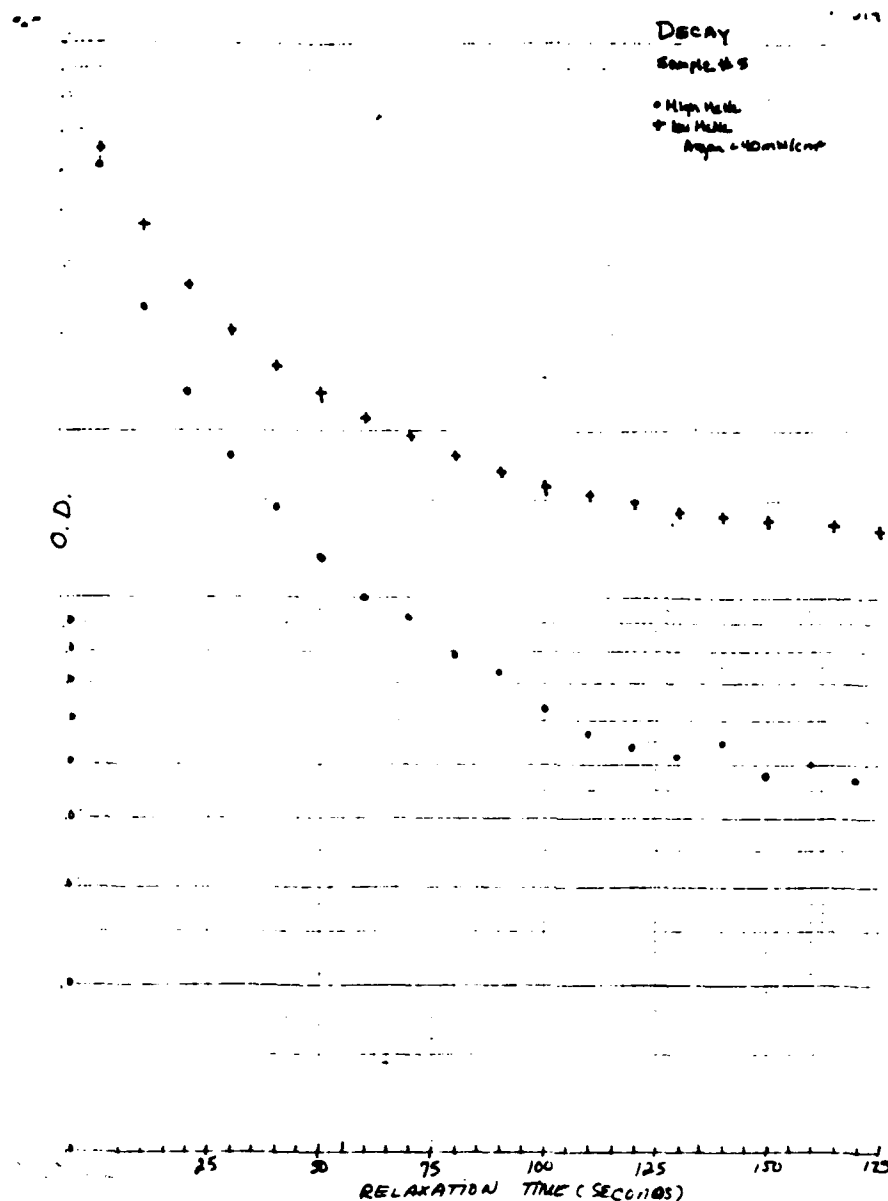order to normalize out the noise factor.

10

Figure 6 - Decay behavior of the optical density at the helium
neon laser wavelength for photochromic sample #5 versus time
measured after illumination with low power argon laser light.
Sample temperature was 90 degrees C. Here two different helium
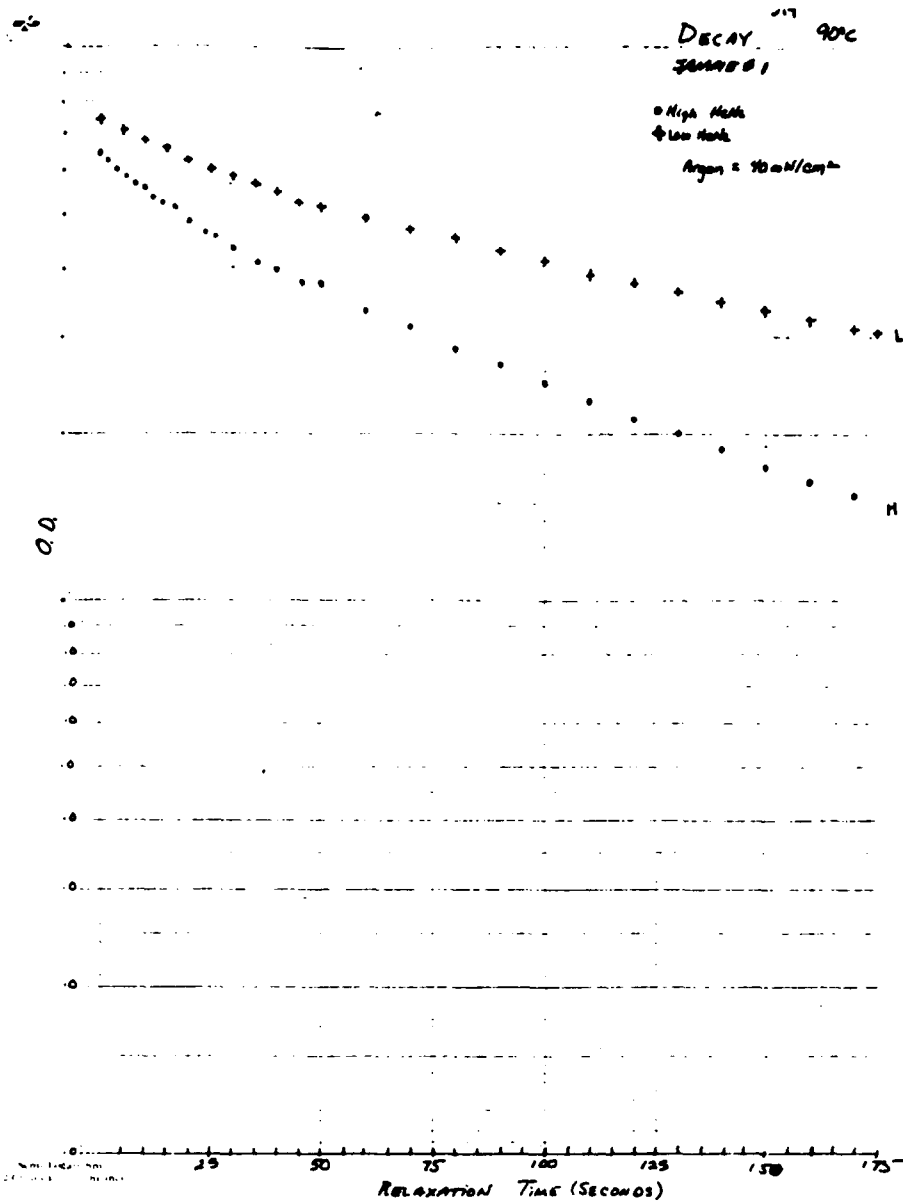neon laser reading intensities were used.

Figure 7 - Decay behavior of the optical density at the helium
neon laser wavelength for photochromic sample #1 versus time
measured after illumination with low power argon laser light.
Sample temperature was 90 degrees C. Here two different helium
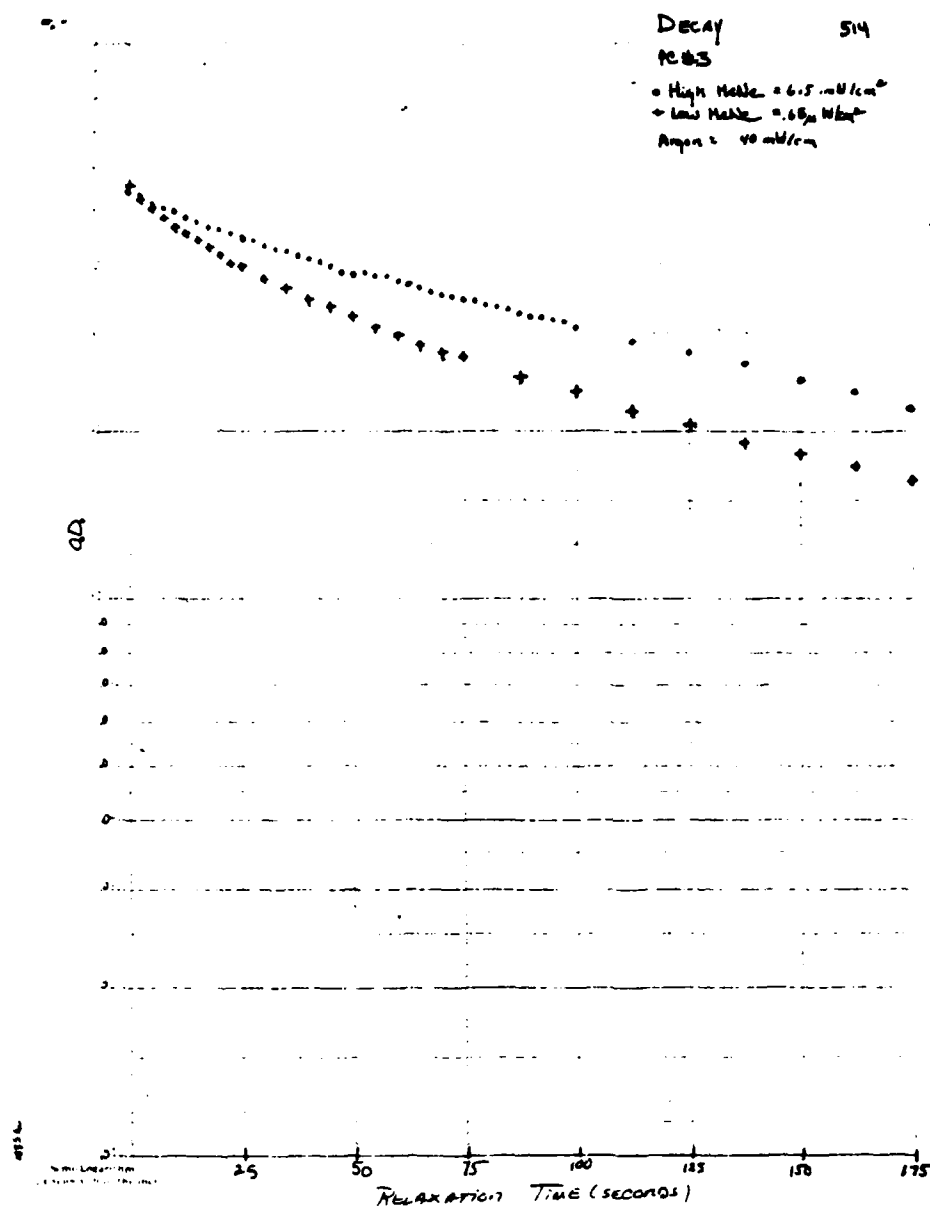neon laser reading intensities were used.

Figure 8 - Decay behavior of the optical density at the helium
neon laser wavelength for photochromic sample #3 versus time
measured after illumination with low power argon laser light.
Sample temperature was 90 degrees C. Here two different helium
neon laser reading intensities were used.

13

Secondly, we found that the samples are affected by long time exposure to room lights. Once this was recognized, samples were kept in the dark for 3 days prior to exposure to insure that the samples were completely in their original ground states.

To summarize, we have measured different erasure time effects in our three samples. The decay time is decreased with increasing sample temperature. In addition, the decay rate is affected by increasing helium neon laser reading intensity.

After sufficient experimentation to identify some of these qualitative features, we discussed our results with Dr. Nori Chu of American Optical Corp.

He verified our conclusions and informed us that the composition of the solid samples is different from that of the liquid samples reported by Goodwin and Mottola (2). He said that his photochromic samples can exist in two species. The first is mercury bis-dithizonate in which the mercury is bound to two ligands. This is the same as reported by Goodwin. However, a second phase can exist called phenyl mercury dithizonate in which only one ligand is bound to the mercury. This second species is characterized by faster kinetics than the first.

In his method of fabricating samples of high optical quality, he could not control the fraction of these two species. However, he can utilize a casting technique in which either species can be obtained but where the optical quality might be sacrificed. We hope to work with these samples in an extension of this work.

Therefore, our present results now make sense and can be explained by the fact that our samples have different concentrations of the two species with different kinetics.

Next we will consider our experiments measuring the writing time behavior of the samples. In these experiments, we also noticed a dependence on the helium neon laser intensity for low writing powers. This dependence on the helium neon reading intensity will be discussed in the last chapter along with our other conclusions.

IV. WRITING SPEED EXPERIMENTS

We next investigated the writing characteristics of the samples
as a function of time under different argon laser writing powers.
Several introductory comments are in order.

First, the writing time should decrease as the argon laser
intensity was increased. However, if the writing time is long
enough compared with the decay times of the photochromic species,
then the overall writing time should be extended due to the
competition between the two processes. This competition is
eliminated as the writing intensity increases.

In addition, our experiments with the different power helium neon
reading beams indicate that when the writing time is comparable
to the decay time, then the writing time should be affected by
the helium neon laser intensity. This effect will also diminish
as the writing intensity increases.

We expect the writing time to be temperature dependent since the
decay times decrease with increasing temperature.

Finally, we expect a variation in the writing speed for different
samples since they have different compositions of fast and slow
decay species.

In our monthly reports, we spent a great deal of time discussing
specific experiments at various writing intensities. In this
final report, we will confine ourselves to a summary of results
obtained with a range of writing intensities.

Figure 9 shows a log plot showing the exposure time required to
obtain various optical densities for sample #1 as a function of
writing intensity using the 488.0 nm laser wavelength and a
sample temperature of 90 deg C. The particular optical densities
were arbitrarily chosen at O.D. = 1.0, 0.3, and 0.1. As
hypothesized, there is a linear relation except at low
intensities where, as expected, the writing times are lengthened
due to the competition between decay times and writing times.
Figure 10 shows the same plot on an expanded scale.

Figure 11 shows the same plot with a different sample temperature
of 10 deg C. In this case, the curves are more linear. This is
expected since the decay times are slower at lower sample
temperatures and there is not as much competition with the
writing speeds. In general, the samples wrote more slowly at
lower temperature.

Figure 12 again shows the same results as in Figure 11 but on an
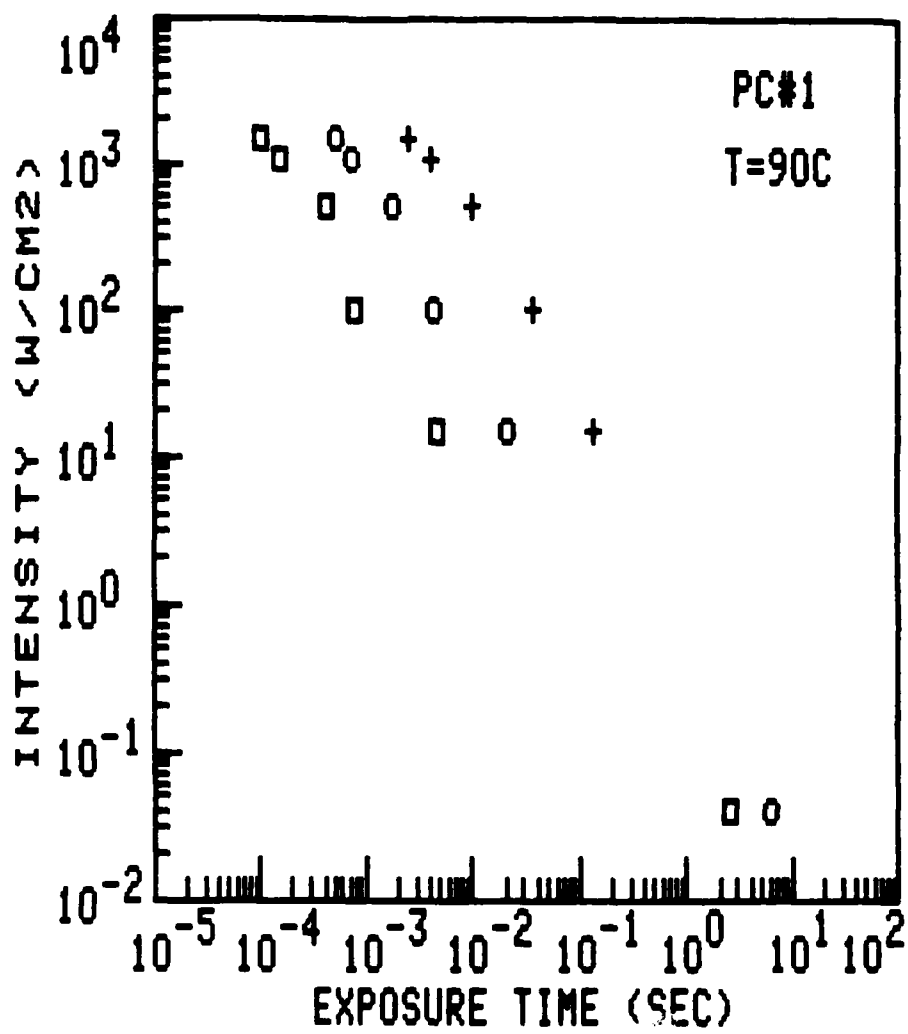expanded scale.

15

Figure 9 - Exposure time required to reach various optical densitities as a function of laser intensity at 488.0 nm for sample #1 at a temperature of 90 degrees C. Open squares correspond to OD=0.1, open circles OD=0.3, crosses OD=1.
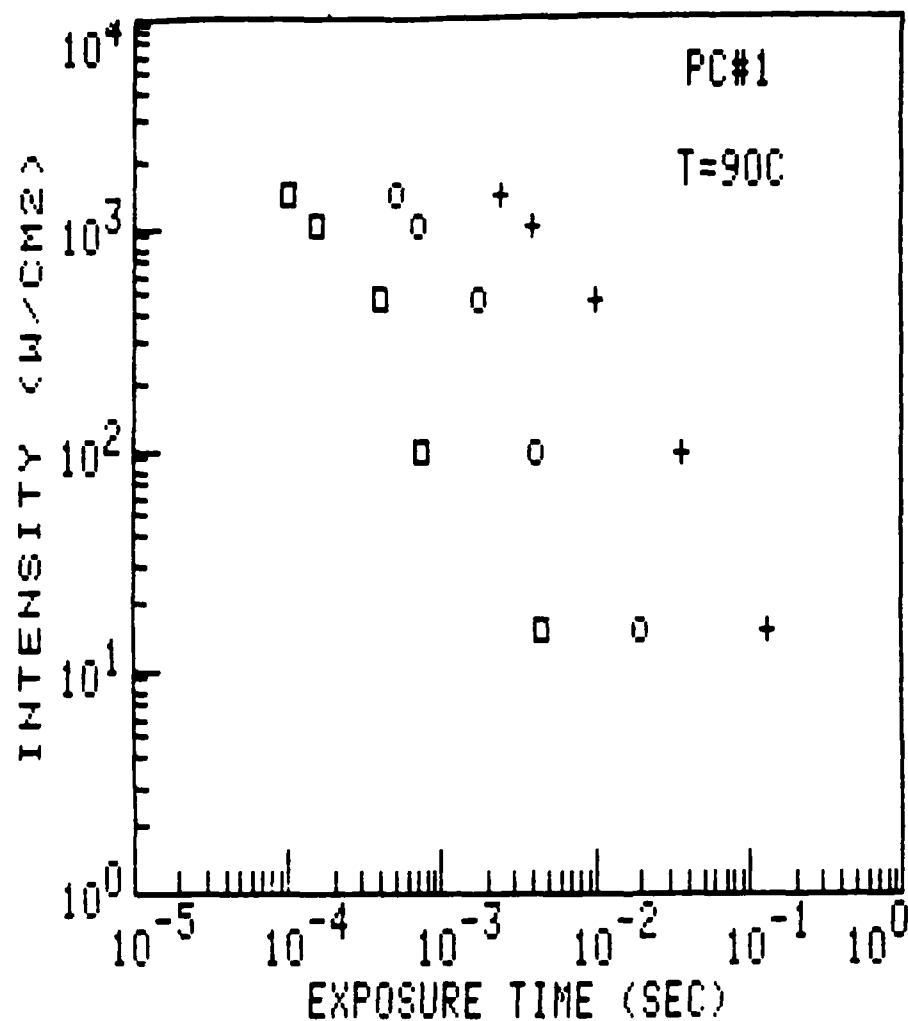
16

Figure 10 - Exposure time required to reach various optical densities as a function of laser intensity at 488.0 nm for sample #1 at a temperature of 90 degrees C. Open squares correspond to OD=0.1, open circles OD=0.3, crosses OD=1.

17

Figure 11 - Exposure time required to reach various optical
densitities as a function of laser intensity at 488.0 nm for
sample #1 at a temperature of 10 degrees C. Open squares
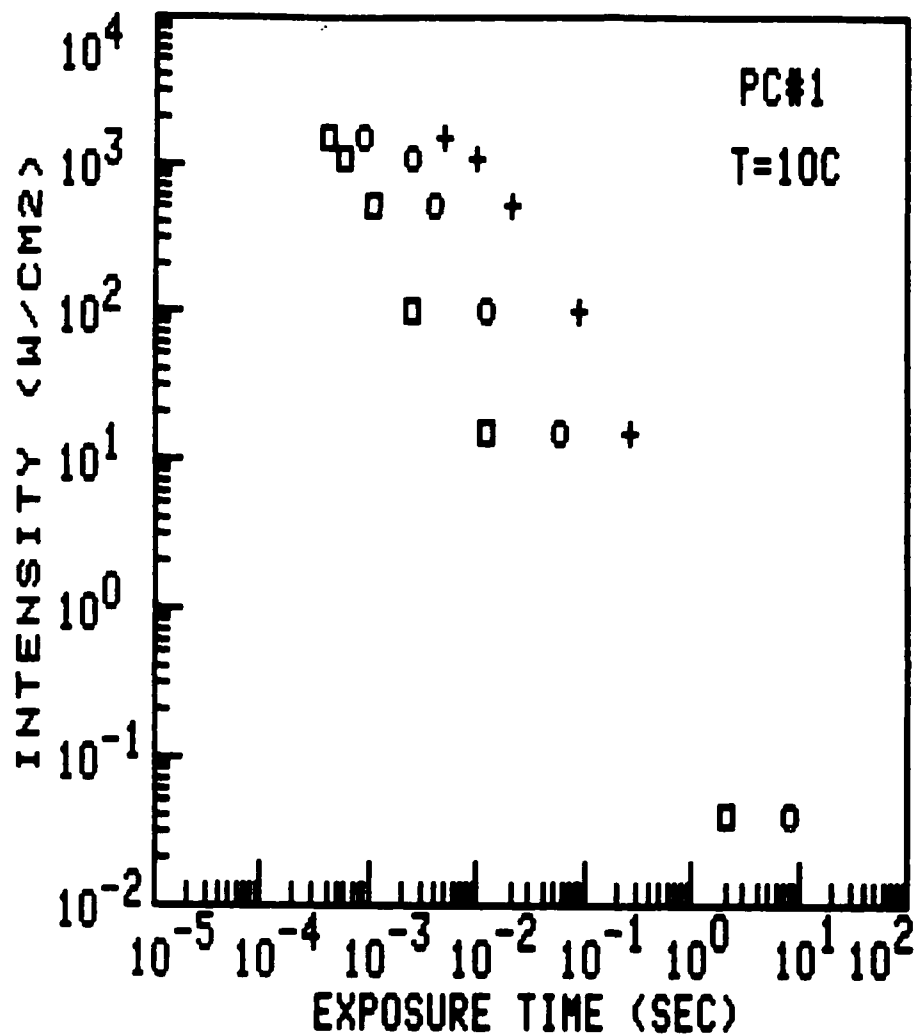correspond to OD=0.1, open circles OD=0.3, crosses OD=1.

18

Figure 12 - Exposure time required to reach various optical densitities as a function of laser intensity at 488.0 nm for sample #1 at a temperature of 10 degrees C. Open squares correspond to OD=0.1, open circles OD=0.3, crosses OD=1.
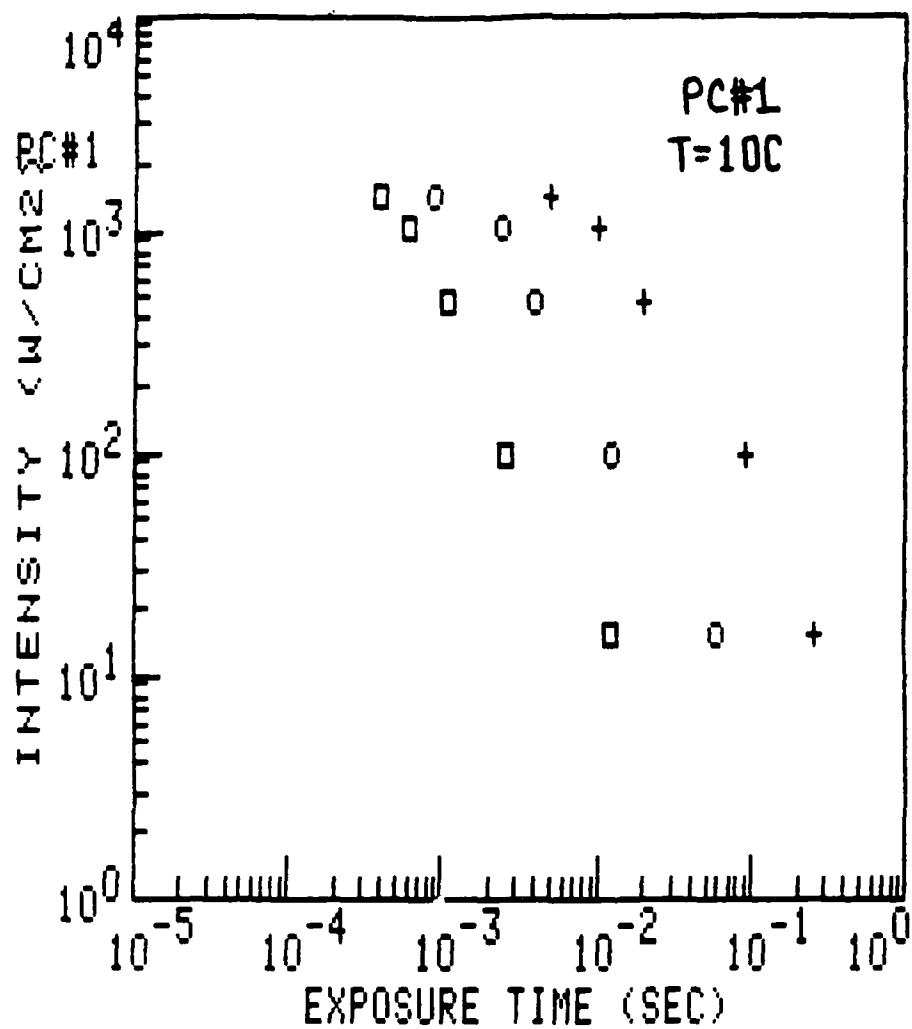
19

Comparison of Figures 10 and 12 shows that the higher sample
temperature results in faster writing speeds.

From the slope of these curves, we can obtain the exposure energy
required to reach a desired optical density and results are shown
in Figure 13. As expected, the sample is more efficient at high
temperatures and requires less energy to reach a particular
optical density than at lower temperatures.

We expected different results from the other samples due to their
different compositions. One interesting set of experiments
examined the writing time as a function of wavelength at a fixed
laser intensity for the three samples.

Figure 14 shows the optical density of sample #1 at a sample
temperature of 90 deg C as a function of time for a writing
intensity of 15 W/sq cm. We see that the 514.5 nm wavelength is
more effective for writing this sample.

Figure 15 shows the optical density of sample #5 as a function of
time for a writing intensity of 15 W/sq cm at the same sample
temperature. In this sample, there is little difference between
the two writing wavelengths.

However, the results are very different in sample #3. Figure 16
shows the optical density of sample #3 as a function of time for
a writing intensity of 15 W/sq cm. We see that now the 488.0 nm
wavelength is more effective for writing this sample.

Overall, sample #1 was the fastest while sample #3 was the
slowest. This indicates that the SLOW component is more
absorbing at the 488.0 nm wavelength while the FAST component is
more absorbing at the 514.5 nm wavelength. This is a very
significant result!

These results are more dramatic when plotted in a different
format. In Figure 17, the optical densities as function of time
are shown for all three samples under illumination at 488.0 nm.
No significant difference is seen.

However, the same plot is shown in Figure 18 when the laser
wavelength is at 514.5 nm. Here the speed of sample #1 is
clearly seen.

Figure 19 shows the fastest writing speeds which we obtained with
optical density versus writing time for an exposure intensity of
1400 watts/sq cm with sample #1 at temperatures of 90 and 10
degrees C. The OD = 1 point is reached within a few
milliseconds. Note that this experiment was performed at the
laser wavelength of 488.0 nm and that faster speeds are expected
with 514.5 nm laser light. In addition, this shows very fast
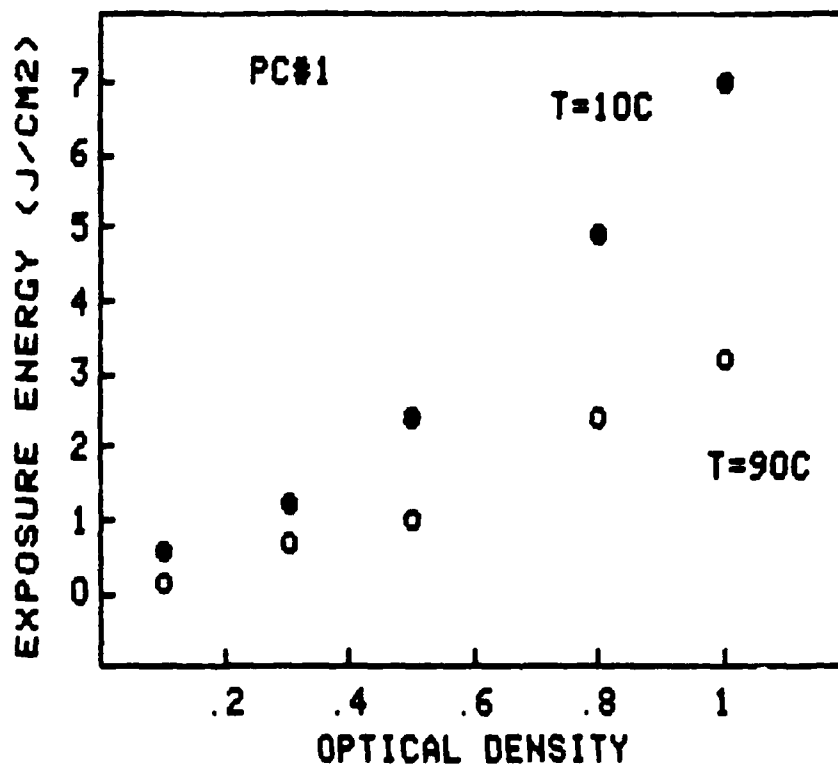writing speeds even at room temperature.

20

Figure 13 - Exposure energy required to reach various optical densities at a laser wavelength of 488.0 nm for sample #1. Graph shows sample tempertures of 10 and 90 degrees C.
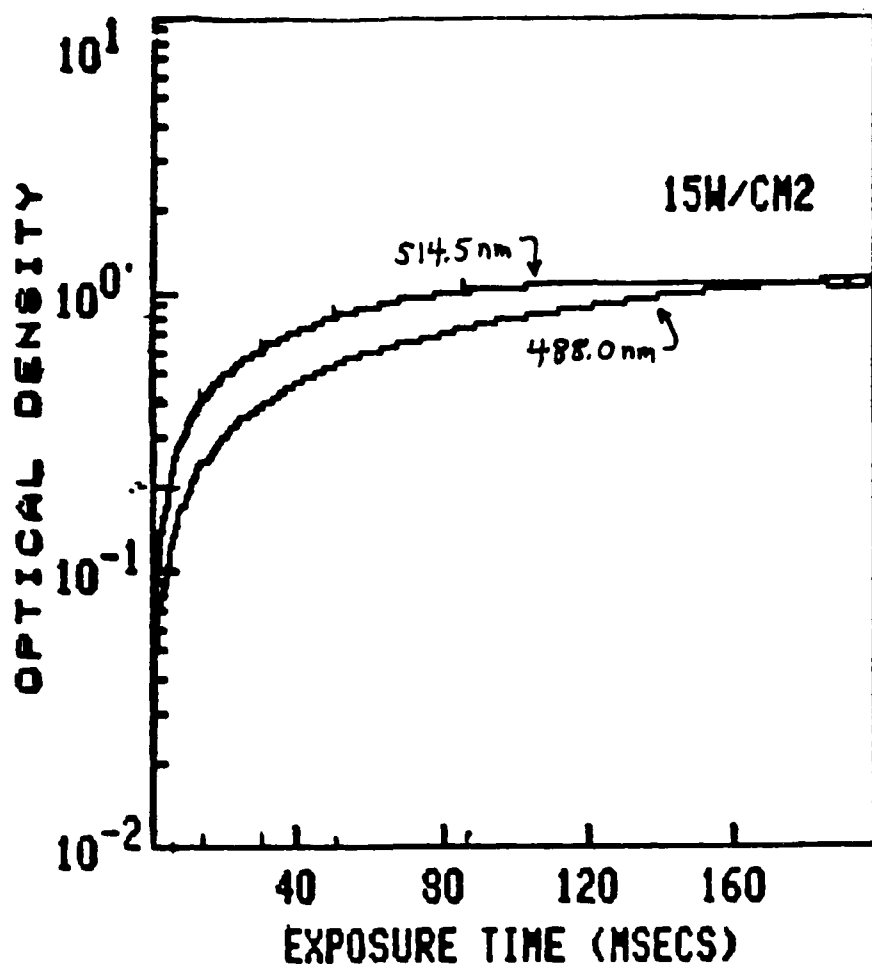
Figure 14 — Optical density as a function of writing time for
sample #1 at a temperature of 90 degrees C and a laser intensity
of 15 W/sq cm for two different laser wavelengths

Figure 15 - Optical density as a function of writing time for sample #5 at a temperature of 90 degrees C and a laser intensity of 15 W/sq cm for two different laser wavelengths
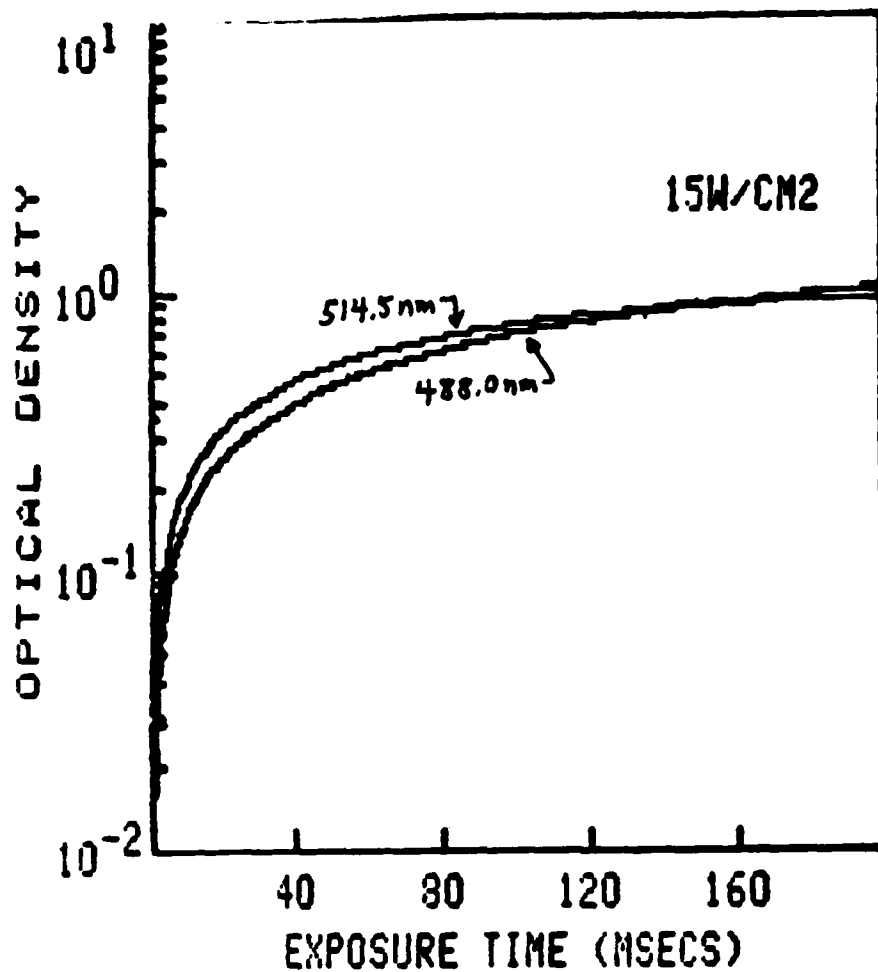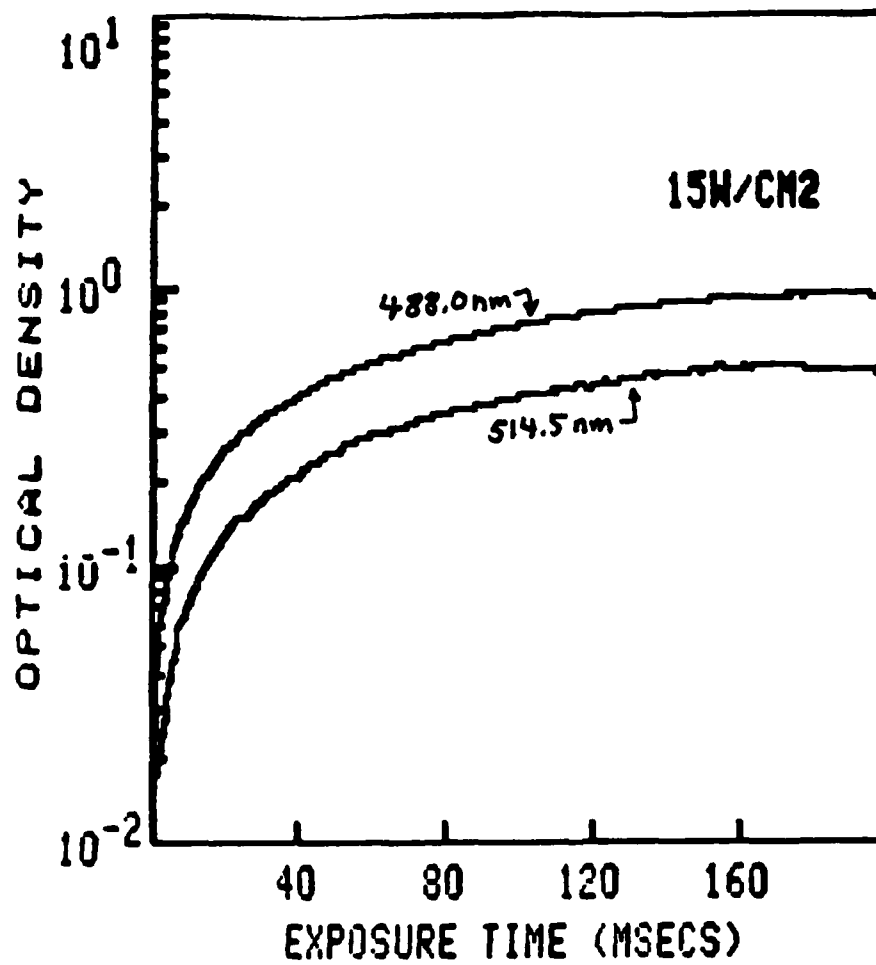
23

Figure 16 - Optical density as a function of writing time for sample #3 at a temperature of 90 degrees C and a laser intensity of 15 W/sq cm for two different laser wavelengths

24

Figure 17 - Optical density as a function of writing time for
samples #1,#3, and #5 at a temperature of 90 degrees C and a
laser intensity of 15 W/sq cm for a laser wavelength of 488 nm.

25
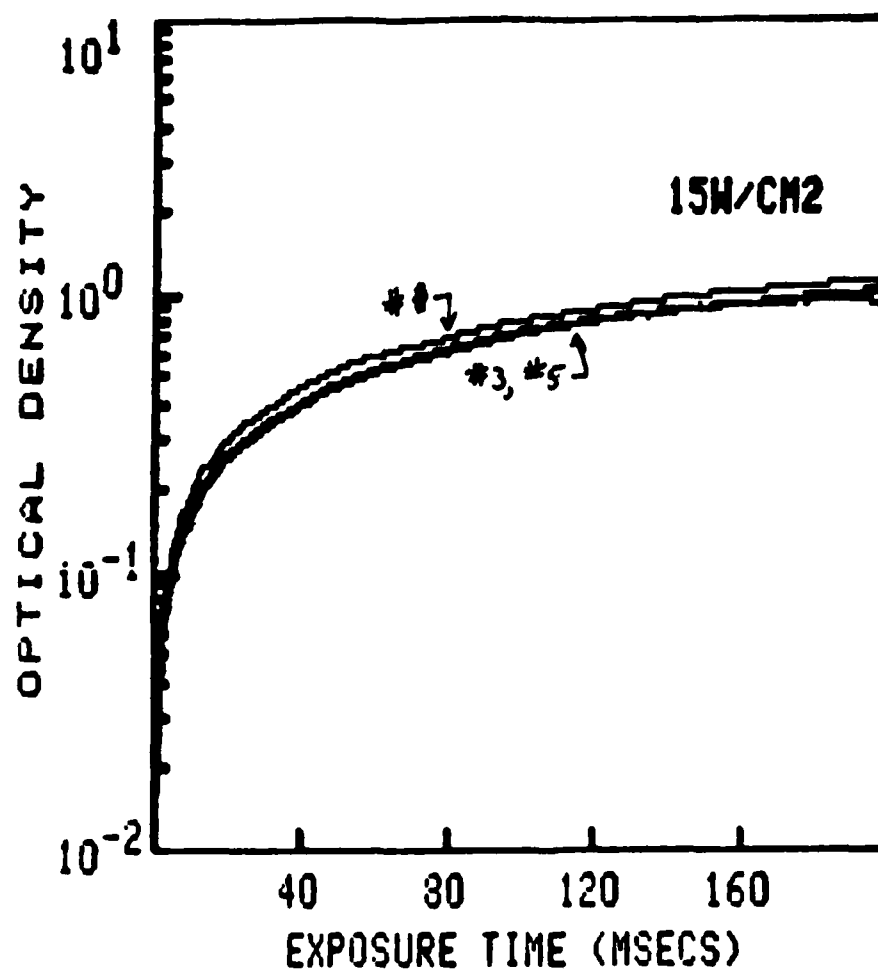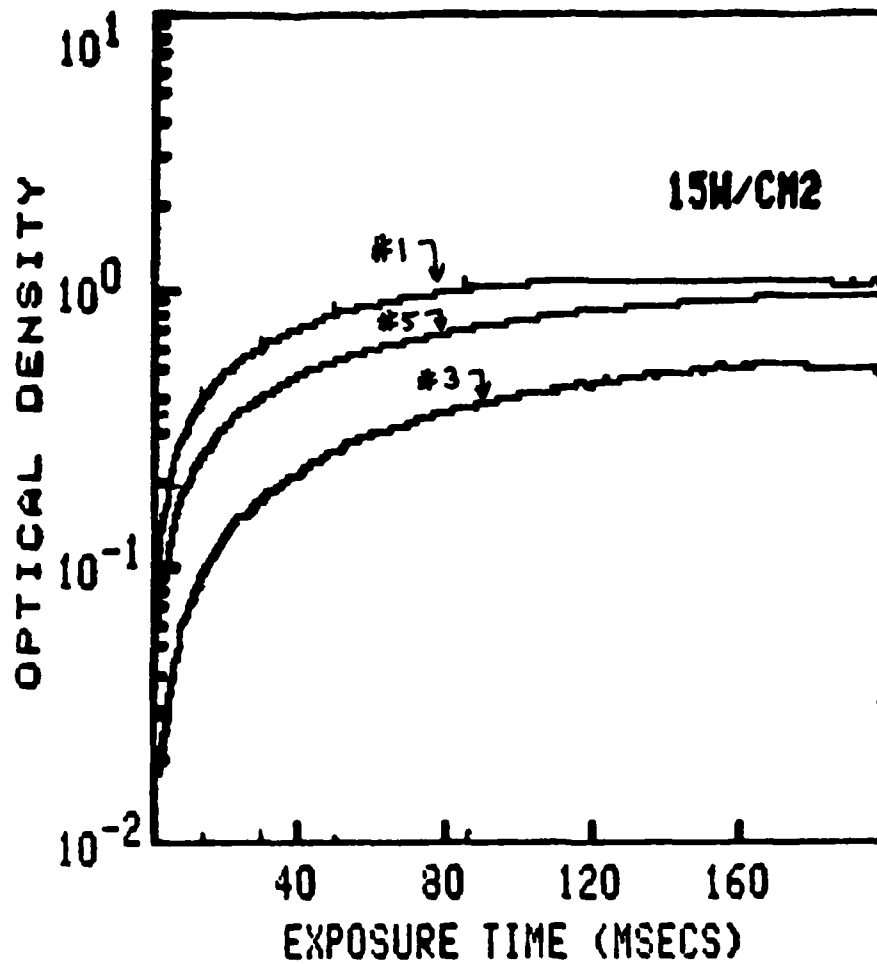
Figure 18 - Optical density as a function of writing time for
samples #1,#3, and #5 at a temperature of 90 degrees C and a
laser intensity of 15 W/sq cm for a laser wavelength of 514.5 nm.
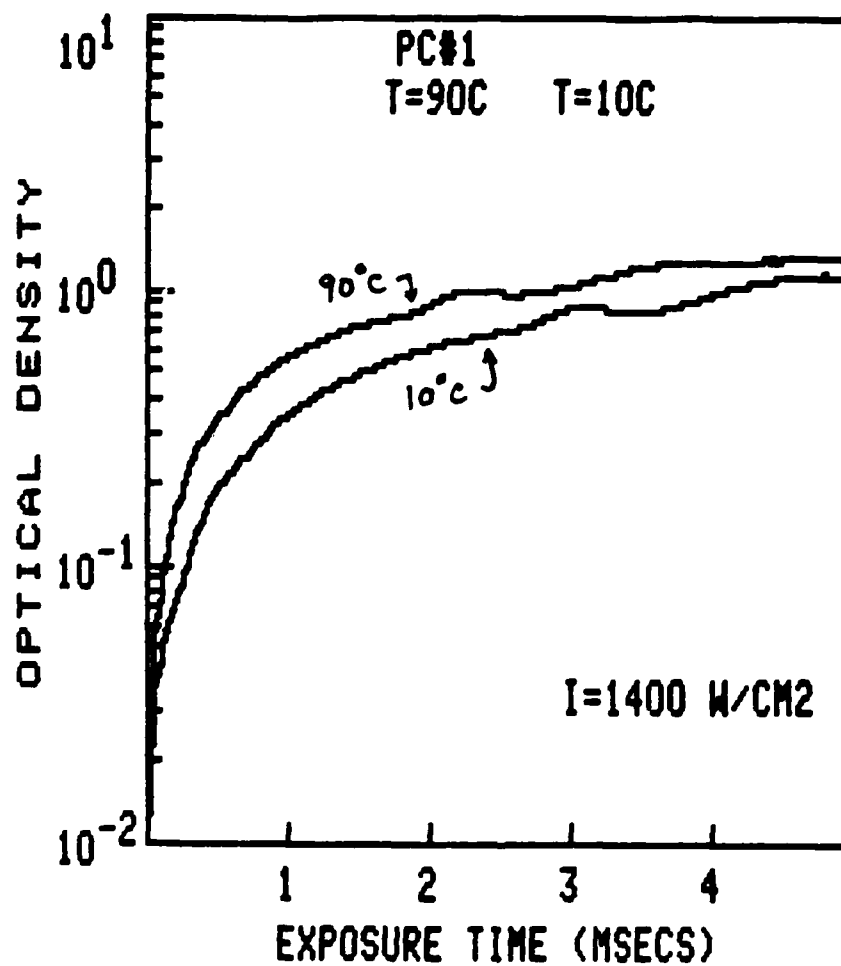
26

Figure 19 - Optical density versus writing time for laser
exposure of 1400 W/sq cm with sample #1 at sample temperatures of
90 and 10 deg C. Writing laser wavelength is 488.0 nm.

Finally, the effects of the helium neon laser intensity on the
writing speed were examined. We already reported experiments on
the decay time in which the decay was enhanced with increased
helium neon illumination. Under low intensity writing
conditions, we would expect that the fast decay process would
limit the speed with which this sample becomes opaque.
Therefore, the writing time should INCREASE as the helium neon
laser intensity increases because the decay process is faster.
This is indeed observed and is shown in Figure 20. Note that
this dependence was seen under low writing laser intensities of 5
mW/sq cm. It disappeared as expected at higher writing laser
intensities.

To summarize our observations, we see that the writing speed is
different for the three different samples. Interpretation of
these results is consistent with the assumption that we have two
different photochromic species with different concentrations of
the two species with different kinetics. It is clear that one of
the species has a much faster relaxation time than the other.

We found, as expected, that a certain exposure energy is required
to write on the photochromic materials. The exposure time needed
to write the photochromic to a certain optical density can be
decreased by increasing the intensity of the writing beam and by
increasing the sample temperature.

In each case, the higher temperature sample shows both an
increased initial writing speed and a higher final optical
density compared with the lower sample temperature.

However, sample #1 is more efficient, reaching a higher saturated
optical density more quickly than either of the other two.
Sample #5 is slightly faster than #3.

Finally, we see sample dependent effects in which the writing
process depends on the writing laser wavelength.

The final section of this report will discuss these results in
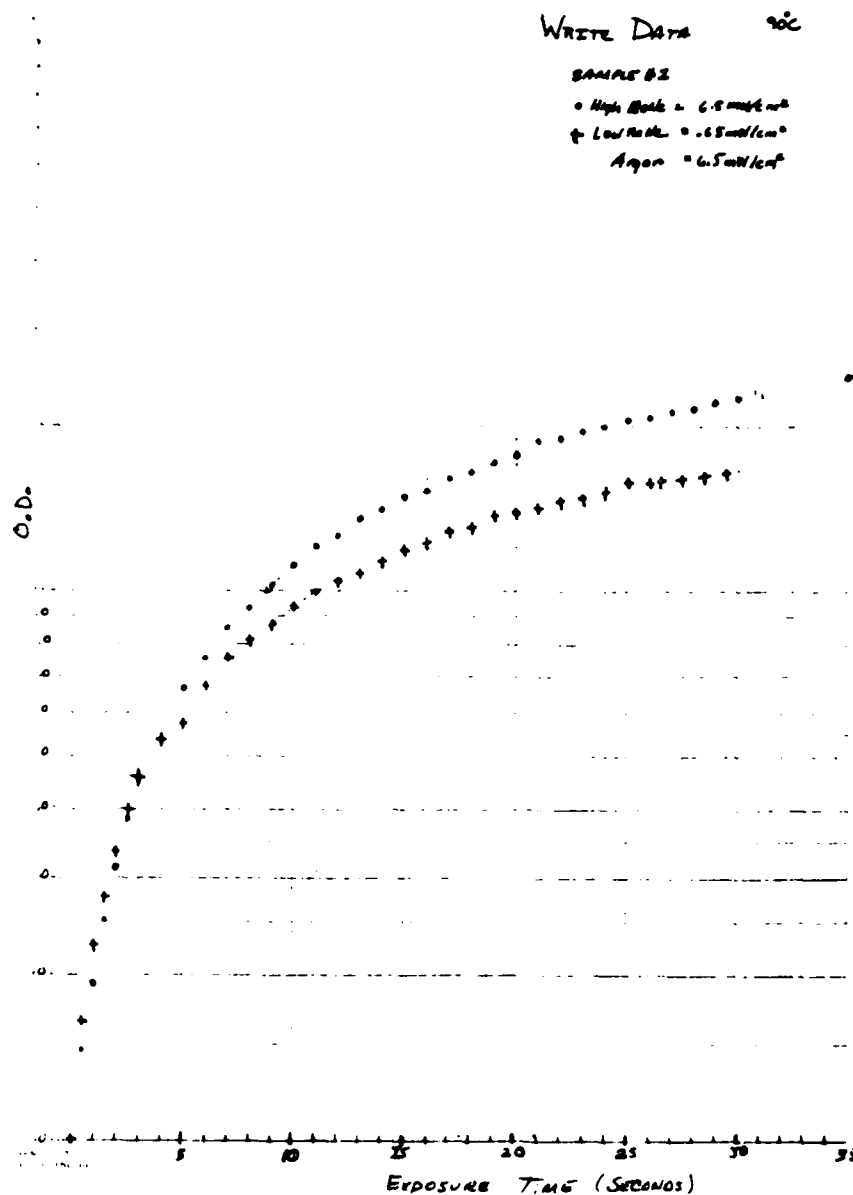the context of the photochromic molecular structure.

28

Figure 20 - Optical density versus writing exposure time for sample #1 at temperature of 90 degrees C using a low power argon writing power. The dots refer to a higher helium neon laser reading power relative to the crosses.

V. CONCLUSIONS

In summary, we have successfully examined the writing and erasure
characteristics of several samples of mercury dithizonate
photochromic films. We are quite excited by the results which
demonstrate an optical recording medium which operates at room
temperature, is inexpensive and reversible, and has a millisecond
response time. Based on these efforts, we feel that we can
exceed this speed.

There are several factors which we believe have affected our
experiments and prevented us from obtaining even faster writing
speeds.

As has been discussed previously, our samples are composed of two
species of mercury dithizonate each with different kinetics.
However, we have no data on either the composition or the
concentrations of these species for any of our samples. This has
prevented us from performing quantitative analyses on our data.
Nevertheless, we can draw some general conclusions. However, we
will first review some of the background of these materials.

Photochromic materials involve the conversion through optical
absorption of one form of a molecule (labeled A) to another form
(labeled B). Both species have different absorption spectra as
shown in Figure 21 for mercury dithizonate (3). In this figure,
the dashed curve shows the absorption spectrum as a function of
wavelength when all of the molecules are in the ground or
unexcited state referred to as the A state. Under optical
excitation in the 440 nm region, there is a conversion to the
excited state B whose spectrum is given by the solid curve.
There is a pronounced increase in the optical absorption for the
red spectral region.

If there are several species of photochromic molecules as in our
case, each is characterized by similar absorption spectra for the
ground state and excited species.

One object of our experiments has been to increase the size and
speed of the red absorption curve. This absorption coefficient
is governed by the number of molecules excited into the B state
denoted by $N(B)$ as well as the absorption cross section for the B
species at the wavelength in question denoted by $S(B)$. Each
species of photochromic molecule will thus be characterized by
its particular values for both of these factors. Since the
wavelength dependence of $S(B)$ cannot be altered, increasing the
absorption for a particular wavelength of red light can only be
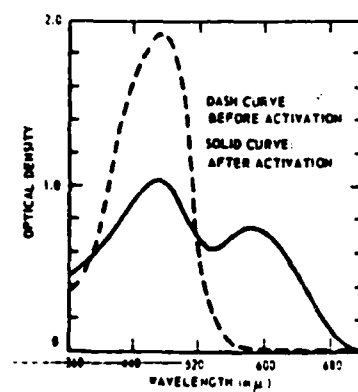accomplished by increasing $N(B)$.

Figure 21 - Absorption spectra of mercury dithizonate
photochromic material. (from Ref 3)

31

The rate of increase in N(B) is directly related to the rate of decrease in the population of the ground state species N(A) and is governed by Beer's Law as

$$dN(A)/dt = -S(A)N(A)F(A)I(B) \qquad Eqn. \ (1)$$

where S(A) is the absorption cross section of the ground state species at the blue-green wavelength, N(A) is the concentration of the photochromic ground state species, F(A) is the quantum efficiency for the ground state species, and I(B) is the intensity of the blue-green writing laser.

This simple expression neglects the existence of additional photochromic species as well as competing decay mechanisms which decrease the excited state population.

Examination of Equation (1) indicates that increasing any of the terms on the right hand side of the equation will increase the writing rate.

Obviously, an increase in the laser intensity will increase the writing rate and this is confirmed by our experiments.

The temperature dependence most probably enters through a variation in the quantum efficiency. The physical basis of this will be considered below.

The wavelength dependence of the writing speed which was shown in Figure 14 is due to different absorption cross sections for the two species which are present in our samples. This will be covered in more detail later.

Finally, changing the concentration of the particular photochromic species will affect the writing speed.

As mentioned earlier, there are complications to this simple treatment. The competition between the decay and writing processes was amply illustrated earlier in this report as well as in the progress reports. However, under the high writing intensities which are of greatest interest, these effects are negligible.

The sensitivity in writing speeds due to the helium neon laser intensity at low argon writing powers is most probably due to the photochromism of the excess free ligand, dithizone, whose absorption spectrum has a red component. While of some interest, it does not enter into the high writing speed experiments and will not be discussed further.

In order to further explain our results, we must review the basic structure of the two species of photochromic molecules which are present in our samples.

32

The slower photochromic species is mercury bis-dithizonate whose photochromic mechanism to the excited state involves a cis-trans isomerization about the carbon-nitrogen double bond and a hydrogen exchange between the sulfur and the nitrogen atoms (3). In this case, this molecule is quite large and involves the simultaneous flipping of two sets of molecules resulting in a slower relaxation process.

According to Dr. Chu, the faster species is phenyl mercury dithizonate and has only one dithizonate compound bonded to the mercury atom. Its excitation mechanism also involves a cis-trans isomerization about the carbon-nitrogen double bond. However, in this case, the flipping excitation mechanism only involves one molecule rather than two and the process is faster (3).

Both mechanisms involve a structural deformation of the molecules which are aided by increasing the sample temperature. This mechanism explains the measured increase in the relaxation process with increasing temperature.

There is some evidence showing different absorption spectra for these two photochromic species which might explain our wavelength data. Figure 22 shows the ground state absorption spectrum for primary mercury (II) dithizonate mixed in various solvents (4). The absorption curves all peak near 495 nm.

By contrast, Figure 23 shows the ground state absorption spectrum for TWO other forms of mercury dithizonate (4). Curves 1-4 show the spectrum for secondary mercury (II) dithizonate in various solvents. In this case, the absorption spectrum is peaked further in the green although it is somewhat sensitive to the choice of solvent.

Curve 5 shows the absorption spectrum for primary mercury (I) dithizonate which is also peaked near 490 nm.

The literature is very poor in correlating the two species referred to by Dr. Chu and these three species. The different writing speed experiments versus wavelength reported earlier demonstrate that we have the two forms of mercury (II) dithizonate. However, this is a subject for further explorations.

In conclusion, several aspects of our experimental results are therefore explained. The increased writing speed at higher temperatures is caused by an increase in the quantum efficiency term.

In addition, choice of the 514.5 nm writing beam optimizes the population increase for the faster species. If the sample has a greater fraction of this species, they will be preferentially .np
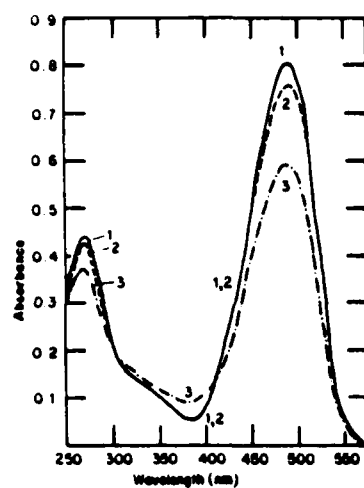
33

STUDIES WITH DITHIZONE. XXIII

Figure 22 - Absorption spectra of primary mercury (II)
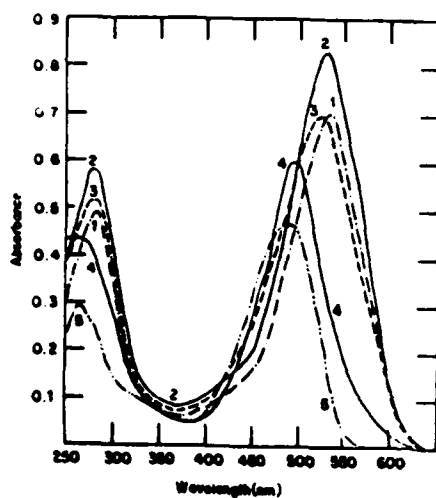dithizonate in various solvents. (from Ref 4)

491

Figure 23 - Absoprption spectra of secondary mercury (II)
dithizonate (curves 1-4) and primary mercury (I) dithizonate
(curve 5) in various solvents.(from Ref 4)

excited by this wavelength. For samples with less of this species, the wavelength dependence will not be as dramatic.

Overall the decay or writing speed of the particular photochromic sample depends on the relative concentrations of these two species. We speculate that sample #1 consists of a greater fraction of the phenyl mercury dithizonate while sample #3 contains a greater fraction of mercury bis thizonate.

Our experiments successfully showed the decrease in writing times as the laser writing intensity increased. However, several limiting factors became apparent.

One is sample damage. If the sample starts to melt, a hole in the sample will allow more helium neon laser light to be transmitted. There is some evidence for this in our data where the optical density is starting to turn down at high exposure energies.

One technique for reducing this damage is to reduce the substrate temperature. While previous results showed a dramatic slowing of the writing time with lower temperature, this effect is minimized at higher writing intensities by the local heating of the sample substrate as shown in Figure 20.

The sample thickness appears to play a role. With a thinner sample, the heat capacity of the sample will be lower. Therefore, the induced heating will be more effective during the writing time. As a result, a lower initial sample temperature can be used and the writing beam will be more effective in increasing the sample temperature during the writing time period. This is also a topic for further investigation.

In conclusion, we have successfully demonstrated the fast writing speed capabiity of mercury dithizonate under intense argon laser illumination demonstrating the potential for this material in optical data processing applications. Much of the work was complicated by unknown sample composition. We have submitted a renewal proposal which specifically addresses this issue since samples can be obtained with known concentrations of particular photochromic species. With our greater understanding of the experimental problems in these measurements, we are confident that substantial progress can be made in quantifying the dynamics of these materials, substantially increasing the writing speed, and optimizing a sample composition for optical data processing applications.

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